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Towards imaging the spatial distribution of geochemical heterogeneities and arsenic sources

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ABSTRACT: We propose a methodology to image the spatial distribution of reactive minerals in the subsurface based on distributed sensor measurements of water quality parameters coupled with forward and inverse reactive transport modeling. We focus on kinetic oxidative dissolution of pyrite and As-bearing pyrite minerals and illustrate the potential of the methodology in synthetic modeling applications at different scales, as well as in laboratory flow-through experiments in 1-D column setups and 2-D flow-through chambers.

1 INTRODUCTION

The spatial distribution of physical and chemical heterogeneities is critical in many subsurface applications. For instance, the location of reactive minerals is a primary factor controlling the fate and transport of organic and inorganic pollutants in groundwater. The latter include geogenic contamination causing the release of heavy metals and metalloids such as arsenic. A number of studies have focused on using hydrologic measurements and inverse modeling techniques to image physical heterogeneity and the spatial distribution of hydraulic conductivity. However, the applications of such approaches to water quality and reactive transport problems are rare.

In this study we focus on oxidation of pyrite and As-bearing pyrite. This process is of key importance in many natural settings as well as in engineering applications such as managed aquifer recharge.

2 METHODS

2.1 Flow-through experiments

We studied the oxidative dissolution of pyrite in different experimental setups, including batch systems, 1-D column setups and 2-D flow-through chambers. Measurements of water quality parameters such as pH, dissolved oxygen, iron and sulfur were useful to formulate and constrain pyrite dissolution kinetics. In particular, spatially-distributed measurements of

dissolved oxygen in the 1-D and 2-D setups were instrumental for imaging pyrite inclusions. Non-invasive optode sensors along the column setups and at different cross sections in the 2-D system allowed us to measure oxygen transport and consumption at high spatial resolution (2.5 mm spacing). A schematic illustration of the experimental setup with two pyrite inclusions is provided in Figure 1.

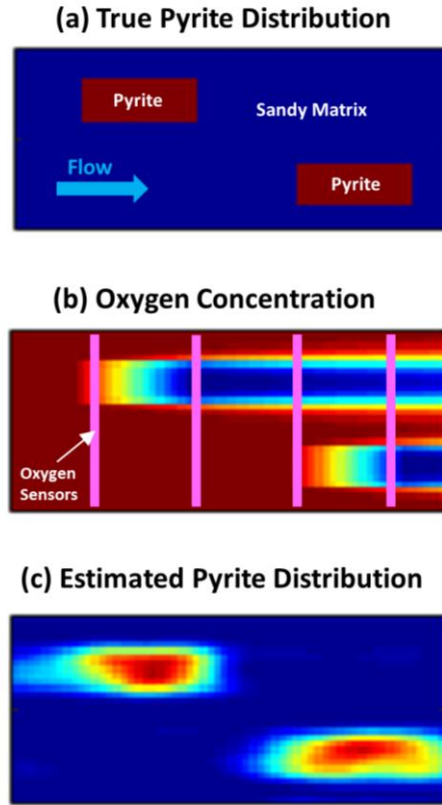


Figure 1. Schematic illustration of the 2-D experimental setup with two pyrite inclusions (a), location of distributed sensor measurements (b), and results of the inverse reactive transport simulations (c).

2.2 Reactive transport modeling

A reactive transport network, including the kinetics of pyrite and As-bearing pyrite oxidative dissolution, was implemented in PHT3D that served as the forward reactive transport simulator. The forward model was applied for synthetic simulations at different scales as well as to quantitatively interpret the laboratory results.

As inverse model, we used the Principal Component Geostatistical Approach (PCGA, Kitanidis & Lee, 2014) to determine the spatial distribution of pyrite and As-bearing pyrite. The PCGA approach was used in combination with the developed forward model and with a limited number of dissolved oxygen observations.

3 RESULTS AND DISCUSSION

The results of the laboratory experiments performed in a number of columns with different size, concentration and number of pyrite inclusions, as well as the outcomes of the 2-D flow-

through experiments are presented by Battistel *et al.* (2017). Here we show some of the results of the synthetic applications at the field scale. Figure 2a shows a 2-D cross section of a shallow aquifer with randomly distributed inclusions of As-bearing pyrite. The inversion was based on synthetic oxygen data, mimicking measurements in multilevel observation wells with 4 m spacing along the flow path. The results show the capability of the approach to correctly locate the As-bearing pyrite inclusions as well as their concentration. The outcomes show a good performance of the inversion also in presence of significant measurement errors (Fig. 2b and 2c).

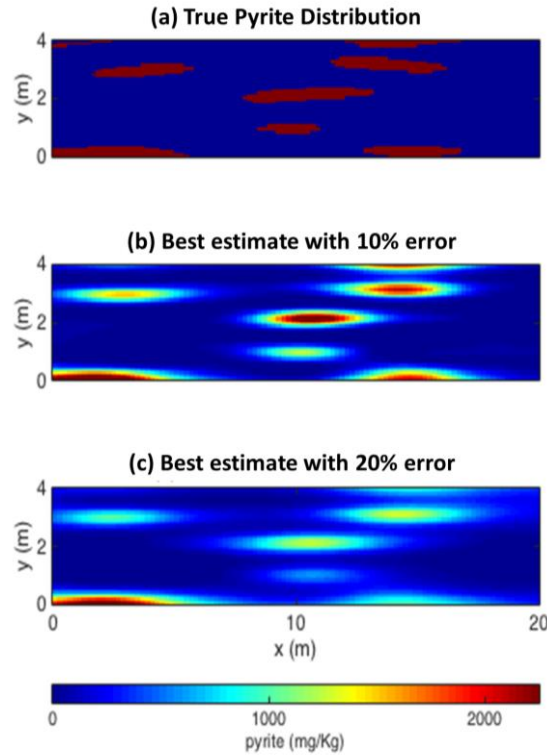


Figure 2. Maps of randomly distributed As-bearing pyrite: true distribution (a) and best estimate in case of 10% error (b) and 20% error (c) in DO measurement (modified from Fakhreddine *et al.*, 2016).

Figure 3 shows the outcomes of simulations performed in the true geochemically heterogeneous aquifer as well as in the best estimate fields obtained with the proposed inverse method. The inverse results show the capability to reproduce the multiple plumes of dissolved arsenic in the heterogeneous domain. An increase in the oxygen measurement error still allows representing the main arsenic plumes, however, the small scale As-sources were more difficult to capture.

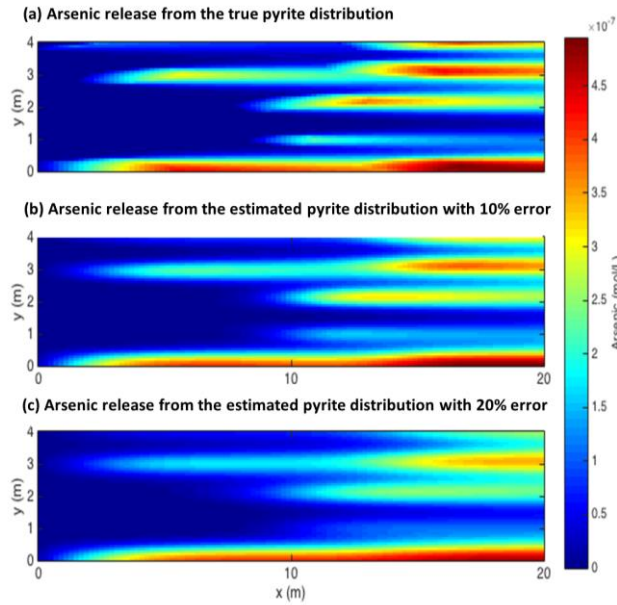


Figure 3. Arsenic plumes in the geochemically heterogeneous aquifers for the true As-bearing pyrite distribution (a), and for the best estimates (b and c) obtained with the proposed inverse modeling approach (reprinted from Fakhreddine *et al.*, 2016, with permission from Elsevier).

4 CONCLUSIONS

We proposed an inverse method for tomography of geochemical heterogeneity. The investigation was focused on imaging the spatial distribution of pyrite and As-bearing pyrite based on spatially distributed oxygen measurements. The methodology proposed is flexible and can be extended to different processes, reactive minerals, as well as distributed measurements of other water quality parameters.

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